IAP20 Rec'd PCT/PTO 09 MAY 2006

Amendment
(under PCT Article 11)
(Partial translation)

To the Commissioner of the Patent Office

- 1. Case number of International Application PCT/JP2004/016915
- 2. Applicant(s)

Name TEIJIN LIMITED

Address 6-7, Minamihommachi 1-chome, Chuo-ku, Osaka-shi, OSAKA 541-0054 JAPAN

Nationality Japan Residing at Japan

3. Patent Attorney

Name (8060) OHSHIMA Masataka

Address OHSHIMA PATENT OFFICE, Fukuya Bldg., 3, Yotsuya 4-chome, Shinjuku-ku, TOKYO 160-0004 JAPAN

- 4. Document(s) to be amended Description and Claims
- 5. Contents to be amended (as per attached sheets)
- (1) "0.001 to 2 μ m" at line 24, page 5 of the original Description (corresponding to line 8, page 17 of the English translation of the Description) was amended to "0.01 to 1 μ m".
- (2) "a stabilization treatment to" at line 3, page 6 of the original Description (corresponding to line 31, page 8 of the English translation of the Description) was amended to "a stabilization treatment by infusibilization in a gas flow to".
- (3) "0.001 to 2 μ m, preferably" at line 9, page 7 of the original Description (corresponding to line 22, page 10 of the English translation of the Description) was deleted.
- (4) "0.001 μ m" at line 11, page 7 of the original Description (corresponding to line 24, page 10 of the English translation of the Description) was amended to "0.01 μ m".
- (5) "2 μ m" at line 12, page 7 of the original Description (corresponding to line 27, page 10 of the English translation of the Description) was amended to "1 μ m".
- (6) "The stabilization can be carried out by in a gas flow is preferred." at lines 17 to 18, page 13 of the original Description (corresponding to line 32, page 19 to line 2, page 20 of the

English translation of the Description) was amended to "The stabilization can be carried out by infusibilization in a gas flow from the viewpoint of productivity.".

- (7) "is preferred" at line 19, page 13 of the original Description (corresponding to line 2, page 20 of the English translation of the Description) was deleted as above (6).
- (8) "0.001 to 2 μ m" at line 2, page 33 of the Claim 1 (corresponding to line 4, page 49 of the English translation of Claims) was amended to "0.01 to 1 μ m".
- (9) Claim 2 at line 5, page 33 (corresponding to lines 6 to 7, page 33 of the English translation of Claims) was cancelled.
- (10) "a stabilization treatment to" at line 23, page 36 of the Claims (corresponding to line 25, page 53 of the English translation of Claims) was amended to "a stabilization treatment by infusibilization in a gas flow to".

6. Attached Sheets:

new pages 5, 6, 7 and 13 of the Japanese PCT Description (corresponding to pages 8, 10, 19 and 20 of the English translation) and

pages 33, 36 and 36/1 of the Japanese PCT Claims (corresponding to pages 49, 53 and 53/1 of the English translation)

fabric of the present invention.

5

15

20

25

30

Still another object of the present invention is to provide a composite material using the above nonwoven fabric of the present invention.

Still another object of the present invention is to provide a metal-carrying nonwoven fabric using the above nonwoven fabric of the present invention and an air cleaning filter using the metal-carrying nonwoven fabric.

Other objects and advantages of the present invention will become apparent from the following description.

According to the present invention, firstly, the above objects and advantages of the present invention are achieved by a nonwoven fabric comprising an aggregate of carbon fibers having a fiber diameter of 0.01 to 1 $\mu m\,.$

According to the present invention, secondly, the above objects and advantages of the present invention are achieved by a method for producing a nonwoven fabric comprising an aggregate of carbon fibers, comprising:

(1) a step of forming an aggregate of precursor fibers from a mixture comprising 100 parts by weight of thermoplastic resin and 1 to 150 parts by weight of at least one thermoplastic carbon precursor selected from the group consisting of pitch, polyacrylonitrile, polycarbodimide, polyimide, polybenzazol and aramid in accordance with a melt blow method,

(2) a step of forming an aggregate of stabilized precursor fibers by subjecting the above aggregate of precursor fibers to a stabilization treatment by infusibilization in a gas flow to stabilize the thermoplastic carbon precursor in the precursor fibers, (3) a step of forming an aggregate of fibrous carbon

weight of metal compound carried on carbon fibers which constitute the nonwoven fabric or crushed material thereof.

5 Brief Description of the Drawings

10

Fig. 1 is a drawing showing a photograph of the surface of a nonwoven fabric comprising carbon fibers obtained by the operation of Example 1 which has been taken by a scanning electron microscope ("S-2400" of Hitachi, Ltd., photographing magnification: 5,000x).

Fig. 2 is a schematic diagram showing a cross section of droplet when a water contact angle is measured in accordance with a drop method.

Fig. 3 is a diagram showing the amounts of

15 trichloroethylene decomposed by titanium oxide

carrying filters of Example 5 and Comparative Example

3 in evaluation of catalytic function.

Best Mode for Carrying out the Invention

The nonwoven fabric of the present invention comprises an aggregate of carbon fibers having a fiber diameter of 0.01 to 1 µm, more preferably 0.05 to 0.5 µm. When the fiber diameter of the carbon fibers is smaller than 0.01 µm, the nonwoven fabric has low mechanical strength and is difficult to handle disadvantageously, while when the fiber diameter is larger than 1 µm, the water contact angle becomes smaller than 140°, and for example, when the nonwoven fabric is used as an electrode for a fuel cell, it is difficult to remove produced water disadvantageously.

The following relational expression (1) is preferably established between the fiber length (L) and fiber diameter (D) of the carbon fibers constituting the nonwoven fabric of the present invention.

As conditions for melt-blowing, a discharge die temperature of 150 to 400°C and a gas temperature of 150 to 400°C are suitably used. The gas blowing rate of melt-blowing influences the fiber diameter of the precursor fibers. The gas blowing rate is generally 2,000 to 100 m/s, more preferably 1,000 to 200 m/s. When the mixture of the thermoplastic resin and the thermoplastic carbon precursor is melt-kneaded and discharged from a die, it is preferred that the melt-kneaded mixture be passed through a pipe in a molten state and sent to the discharge die continuously, and transfer time from melt kneading to discharge from the spinneret is preferably within 10 minutes.

In the step (1), it is preferable to form precursor fibers having a fiber diameter of preferably 0.01 to 20 μm , more preferably 0.05 to 10 μm . (5) Step (2)

10

In the step (2) in the production method of the present invention, a nonwoven fabric comprising 20 stabilized precursor fibers is formed by subjecting the nonwoven fabric comprising precursor fibers prepared in the above step (1) to a stabilization treatment to stabilize the thermoplastic carbon precursor in the precursor fibers. Stabilization of the thermoplastic 25 carbon precursor is a step necessary for obtaining a nonwoven fabric comprising carbonized or graphitized carbon fibers. When the subsequent step of removing the thermoplastic resin is carried out without the stabilization step, there occurs a problem such as 30 thermal decomposition or fusion of the thermoplastic carbon precursor.

The stabilization can be carried out by infusibilization in a gas flow from the viewpoint of productivity. The gas component to be used is

preferably a mixed gas comprising oxygen and/or a halogen gas in view of permeability to the above thermoplastic resin, adsorbability to the above thermoplastic carbon precursor and a fact that the gas can make the thermoplastic carbon precursor infusible quickly at low temperatures. Illustrative examples of the halogen gas include a fluorine gas, chlorine gas, bromine gas and iodine gas. Of these, the bromine gas and the iodine gas are preferred, and the iodine gas is particularly preferred. As a specific method of infusibilization in a gas flow, it is preferable to treat the thermoplastic carbon precursor in a desired gas atmosphere at 50 to 350°C, preferably 80 to 300°C, for 5 hours or less, preferably 2 hours or less.

Further, the softening point of the thermoplastic carbon precursor contained in the precursor fibers is significantly elevated by the above infusibilization. The softening point is preferably 40°C or higher, more preferably 500°C or higher, so as to obtain desired carbon microfibers. By carrying out the above method, the thermoplastic carbon precursor in the precursor fibers is stabilized, and an aggregate (nonwoven fabric) comprising the stabilized precursor fibers can be obtained.

25 (6) Step (3)

5

10

15

20

30

In the step in the production method of the present invention, the thermoplastic resin contained in the stabilized precursor fibers is removed by thermal decomposition. More specifically, the thermoplastic resin contained in the stabilized precursor fibers is removed, only a stabilized fibrous carbon precursor is

CLAIMS

- 1. (Amended) A nonwoven fabric comprising an aggregate of carbon fibers having a fiber diameter of 0.01 to 1 $\mu m\,.$
 - 2. (Cancelled)
- 3. The nonwoven fabric of claim 1, wherein the 10 fiber diameter is 0.05 to 0.5 μm_{\odot}
 - 4. The nonwoven fabric of claim 1, having a density of the carbon fibers per unit area of 1 to 1,000 g/m^2 .

15

- 5. The nonwoven fabric of claim 1, having a density of the carbon fibers per unit area of 2 to 500 $\ensuremath{g/m^2}$.
- 6. The nonwoven fabric of claim 1, having a porosity of 60 to 98%.
 - 7. The nonwoven fabric of claim 1, having a porosity of 80 to 98%.

25

- 8. The nonwoven fabric of claim 1, having a porosity of 90 to 98%.
- 9. The nonwoven fabric of claim 1, having a water 30 contact angle of 140 to 155° at 20°C and a relative humidity of 65 to 70%.
 - 10. The nonwoven fabric of claim 1, having a thickness of 5 μm to 2 cm.

- 34. The metal-carrying nonwoven fabric or metal-carrying pulverized material of claim 32, wherein the form of the metal compound carried is a thin film having a film thickness of 1 to 100 nm or particles having a particle size of 1 to 100 nm.
- 35. A filter substrate comprising the metal-carrying nonwoven fabric or metal-carrying pulverized material of claim 32.

10

20

5

- 36. A water treatment or air cleaning filter comprising the filter substrate of claim 35.
- 37. (Amended) A method for producing a nonwoven 15 fabric comprising an aggregate of carbon fibers, comprising:
 - (1) a step of forming an aggregate of precursor fibers from a mixture comprising 100 parts by weight of thermoplastic resin and 1 to 150 parts by weight of at least one thermoplastic carbon precursor selected from the group consisting of pitch, polyacrylonitrile, polycarbodiimide, polyimide, polybenzazol and aramid, in accordance with a melt blow method,
- (2) a step of forming an aggregate of stabilized precursor fibers by subjecting the above aggregate of precursor fibers to a stabilization treatment by infusibilization in a gas flow to stabilize the thermoplastic carbon precursor in the precursor fibers,
- (3) a step of forming an aggregate of fibrous carbon precursor by removing the thermoplastic resin from the aggregate of stabilized precursor fibers, and
 - (4) a step of carbonizing or graphitizing the aggregate of fibrous carbon precursor.

38. The method of claim 37, wherein the fiber